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Quasioptical study of antiferromagnetic resonance in $YFeO_3$ at submillimeter wavelength under high pulsed magnetic fields

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ABSTRACT

Transmission spectra, $T(H)$, of linearly polarized electromagnetic waves through $YFeO_3$, weak ferromagnet, measured at frequencies $\nu = 96–1000$ GHz in long-pulsed magnetic fields ($H||k||c$-axis, Faraday geometry) exhibit strong rotation of the polarization plane near the quasiferromagnetic AFMR as well as low frequency impurity modes. New ascending impurity branch including five lines was observed at high magnetic field (10–30 T) at 96 GHz and 140 GHz in addition to the known low-field descending impurity branch. Behavior of all the impurity modes assigned to transitions in $^6S_{5/2}$ multiplet of Fe^{3+} "impurity" ions in *c*-sites was described self-consistently by one spin-Hamiltonian. A theoretical calculation of dynamical magnetic susceptibility at AFMR and impurity modes and further simulation of transmission spectra made it possible to describe the main features of the observed spectra $T(H)$. It was found that the $T(H)$ behavior is determined at resonances not only by non-diagonal components of the magnetic susceptibility but also by the anisotropy of the dielectric permittivity ($\varepsilon'_{xx} \neq \varepsilon'_{yy}$), i.e. birefringence.

1. Introduction

Rare-earth (R) orthoferrites $RFeO_3$ belong to a wide class of weak ferromagnets exhibiting very interesting magnetic properties related to an existence of different magnetic interactions, spin-reorientation phase transitions, magnetic excitations and many other phenomena (see, for example, [1,2]). Various magnetic resonance modes, both in iron and rare-earth subsystems of $RFeO_3$, were studied by diverse spectroscopic techniques, such as far-infrared spectroscopy [3], neutron scattering [4], Raman scattering [5], microwave magnetic resonance technique [6,7] and quasioptical backward-wave oscillator spectroscopy [8,9].

New possibilities in such investigations could be provided by the quasioptical far-infrared technique combined with high pulsed magnetic fields [10,11]. As a rule, such measurements are performed in Faraday geometry, i.e. for a propagation of the electromagnetic radiation with a wave vector **k** along external magnetic field (**H**), and are accompanied by a rotation of polarization plane and interference effects, especially near magnetic resonance modes (see, for example, [12]). It requires the control of the polarization radiation to account for and interpret these phenomena.

In this work we have used far-infrared lasers and microwave techniques with a quasioptical control of the polarization to study antiferromagnetic resonance (AFMR) and impurity modes in yttrium orthoferrite $YFeO_3$ in high pulsed magnetic fields (up to 40 T) for the Faraday geometry. Besides two AFMR modes (quasiferromagnetic and quasiantiferromagnetic modes, respectively, at $\nu_1 \sim 300$ GHz and $\nu_2 \sim 590$ GHz for 4.2 K [5,8,9]), $YFeO_3$ exhibits at low temperatures ($T < 50$ K) also few impurities modes ν_{imp} at 255-300 GHz at zero magnetic field [13,9]. Magnetic field applied along the *c*-axis results in softening of the impurity modes, which were assigned to magneto-dipolar transitions in $^6S_{5/2}$ multiplet of Fe^{3+} "impurity" ions in *c*-sites [14]. Such behavior of the impurity modes, representing actually electron paramagnetic resonance in internal exchange and external magnetic fields, could be explained by an opposite direction of the internal effective field and the external one which determines a descending impurity branch for fields below the threshold field $H_0 \sim 15$ T [14] and suggests the existence of an ascending impurity branch for $H > H_0$. The search for such high-field impurity brunch was one of the aims of this work.

2. Experimental results

Single crystals of $YFeO_3$ were grown by a floating zone method with radiation heating [15]. The plane-parallel plate of *c*-cut of the orthorhombic crystal (space group D_{2h}^{16}—Pbnm) with a transverse size ~7–8 mm and thickness $d = 0.993$ mm was prepared for the FIR study.

Measurements in a "transmission" scheme were performed by means of far-infrared laser and microwave techniques in pulsed

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magnetic fields up to 40 T oriented along the weak ferromagnetic moment of the crystal **F**||*c*-axis (Faraday geometry). A control of the polarization was carried out by a wire grid polarizer and an analyzer in a quasioptical cell installed inside the pulsed coil (Fig. 1). To bring the radiation into the channel of the pulsed magnet we used the oversized cylindrical waveguide ($D = 6$ mm), made of a thin-walled stainless steel. It is well known that the cylindrical waveguide changes the linear polarization of the far infrared and sub-millimeter radiation into the elliptical one. To obtain the linear polarization of the incident and transmitted radiation we used tungsten wire grid polarizers (wire diameter, grid period and aperture of 20 μm, 60 μm and 7 mm, respectively) located close to the sample. These polarizers are effective within the entire range of frequencies (96–1000 GHz) taking into account transmission characteristics of such polarizers reported in Ref. [16]. The cylindrical waveguide was coupled to the open space by means of two fused quartz lenses (focal length ~10 mm). A loss of radiation intensity due to conversion of propagating modes was less than 50% for each side.

Bulk metallic pieces should not be used during measurements in strong pulsed magnetic fields because of heating and vibrations induced by eddy-currents. The sample holder, including the diaphragm, some lenses and polarizer mountings, collets for cylindrical wave-guides—were fabricated of polyetheretherketone polymer **PEEK**™. Internal and external surfaces of the sample holder were covered by 10 μm thin gold film to prevent the radiation propagation around the sample. We used two diaphragms with aperture 4 and 6 mm. No noticeable difference was observed in the measurements performed with the two diaphragms, thus we may conclude, that the focal spot was not exceeding 4 mm in diameter.

To check the applicability of the quasi-optical scheme for polarization measurements we tested the propagation of the radiation without the sample. The radiation passed to the detector through the crossed polarizer and analyzer did not exceed 2–3%, thus indicating the polarization degree better than 97%.

The sample was placed at the center of the coil where the homogeneity of the magnetic field is 10^{-3} over 10 mm the coil axis.

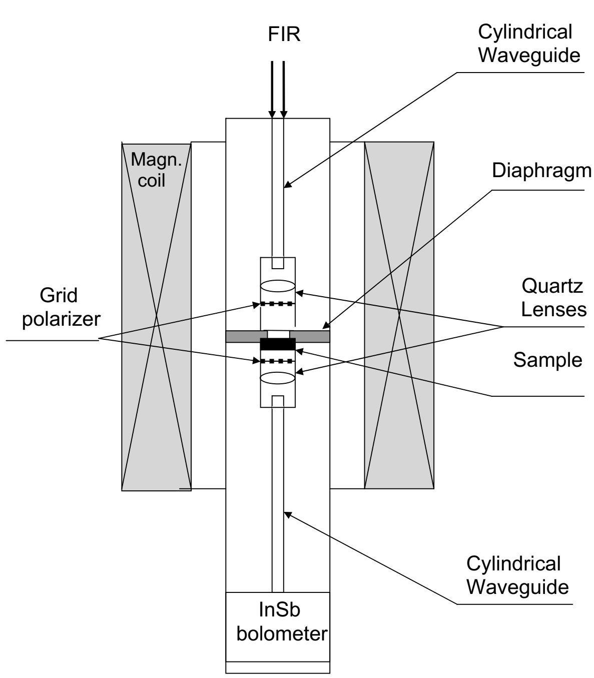

Fig. 1. Scheme of the quasioptical cell installed in a 40 T pulsed coil.

Pulsed magnetic field duration was about 0.2 s, the maximum of the field was reached in 80 ms.

The high frequency measurements (above 250 GHz) were carried out using a gas filled Fabry–Perot cavity optically pumped by a CO_2 laser, while the low frequency studies were performed by means of semiconductor generators (Gunn diode and avalanche diode). A fast InSb bolometer, set out of the coil was used as a detector.

The peculiarity of the quasi-optical configuration is the appearance of interference of the radiation for plane parallel samples, which results in some complexity of transmission spectra. To avoid this interference the wedge-shaped specimens are usually used. In our coil with a small bore diameter it is difficult to use wedge-shaped samples because of parasitic asymmetrical reflections coming from different elements of the quasioptical cell and sample surface, which give rise to multiple standing waves, obstructing the treatment of the transmitted signal. On the other hand plane parallel samples were used in the frequency domain quasi-optical studies [8,9] for quantitative description of spectra and extracting information on a complex permittivity taking into account multiple interferences. In this work we have also used the plane-parallel samples to provide better theoretical treatment of measured spectra.

The main part of the measurements was carried out for crossed analyzer and polarizer geometry in order to study the effects of the polarization plane rotation. The usual approach to measure the polarization characteristics in magnetic fields includes observation of the transmission in crossed polarizer and analyzer, while the polarizer is aligned along a crystallographic axe of the sample. The polarization plane rotation under magnetic field gives rise to a signal at the detector, thus opening a possibility to adjust the system. In our pulsed field experiment, where the adjustment of the measuring system including the radiation source, wave guide and the quasioptical channel, was possible only without the magnetic field, the orientation of the analyzer and polarizer was chosen close to ±45° with respect to the crystallographic *a*- and *b*-axes in the sample plane. In this case a sufficient signal was detected due to the transformation of the linear polarization to the elliptical one owing to a natural birefringence (i.e., anisotropy of permittivity $\varepsilon'_{xx} \neq \varepsilon'_{yy}$). When the orientation of the crossed polarizer and analyzer was along crystallographic *a*- and *b -axes* there were no change of polarization, thus we were not able to adjust the system due to a very weak signal at the detector.

Examples of transmission spectra $T(H)$ near the resonance bands are shown in Figs. 2 and 3 and the dependencies of the observed resonance frequencies as a function of the magnetic field along *c*-axis are displayed in Fig. 4. The increasing high-frequency branch $\nu_1(H)$ represents a known quasiferromagnetic AFMR mode which is excited by the ac magnetic field *h*||*a*- and *b*-axes in the full range of the applied external magnetic fields *H*||*c* [7–9]. The observation of the mode in a crossed analyzer and polarizer configuration suggests a rotation of the polarization plane. The complicated shape of the resonance bands, including multiple picks, indicates significant interference effects inside the line in the plane parallel sample (Fig. 2a–c). We note also a qualitative change of the shape of the resonance bands for different frequencies.

The low frequency branches in Fig. 4 can be identified as impurity modes. The descending branch $\nu_i^-(\mathrm{H})(i = 1, 2, 3, 4, 5)$ including a group of five lines represents the known impurity modes observed in work [14], while the ascending frequencies $\nu_i^+(\mathrm{H})$ is a new impurity branch consisting of also five lines. In the transmission spectra measured without analyzer, the impurity modes look like usual absorption lines while for the crossed analyzer and polarizer geometry the lineshape of the modes changes qualitatively and reveals marked "positive" and "negative" peaks (Fig. 3) resem-